JC07 Rec'd PCT/PTO 15 MAY 2001

FORM PTO-1390 (REV 10-95) U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER WEICKM 10
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. §371		U.S APPLICATION NO. (If known, see 37 CFR §1.5) 09/831825
INTERNATIONAL APPLICATION NO. PCT/EP99/08780	INTERNATIONAL FILING DATE 15 NOVEMBER 1999	PRIORITY DATE CLAIMED 16 NOVEMBER 1998

TITLE OF INVENTION
CRYSTALLINE POROUS SOLIDS, PRODUCTION AND USE THEREOF

APPLICANT(S) FOR DO/EO/US
CHANDRA, Amita, et al.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ■ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. §371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. §371.
3. ☐ This express request to begin national examination procedures (35 U.S.C. §371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. §371(b) and PCT Articles 22 and 39(1).
4. ■ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ■ A copy of the International Application as filed (35 U.S.C. §371(c)(2))
   a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
   b. ■ has been transmitted by the International Bureau.
   c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ■ A translation of the International Application into English (35 U.S.C. §371(c)(2)).
7. ■ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. §371(c)(3))
   a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
   b. ■ have been transmitted by the International Bureau.
   c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
   d. ☐ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. §371(c)(3)).
9. ■ An oath or declaration of the inventor(s) (35 U.S.C. §371(c)(4)).
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. §371(c)(5)).

Items 11. to 16. below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 C.F.R. §§1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 C.F.R. §§3.28 and 3.31 is included.
13. ■ A FIRST preliminary amendment.
    ☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☐ Other items or information:

JC18 Rec'd PCT/PTO 15 MAY 2001

U.S. APPLICATION NO. (if known, see 37 CFR §1.5) 09/831825	INTERNATIONAL APPLICATION NO. PCT/EP99/08780	ATTORNEY'S DOCKET NUMBER WEICKM 10

17. ☒ The following fees are submitted:

BASIC NATIONAL FEE (37 CFR §1.492 (a) (1) - (5)):

		CALCULATIONS	PTO USE ONLY
Search Report has been prepared by the EPO or JPO	$860.00		
International preliminary examination fee paid to USPTO (37 CFR §1.482)	$690.00		
No international preliminary examination fee paid to USPTO (37 CFR §1.482) but international search fee paid to USPTO (37 CFR §1.445(a)(2))	$710.00		
Neither international preliminary examination fee (37 CFR §1.482) nor international search fee (37 CFR §1.445(a)(2)) paid to USPTO	$1000.00		
International preliminary examination fee paid to USPTO (37 CFR §1.482) and all claims satisfied provisions of PCT Article 33(2)-(4)	$100.00		
ENTER APPROPRIATE BASIC FEE AMOUNT =		$860.00	
Surcharge of **$130.00** for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 C.F.R. §1.492(e)).			

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	16 - 20 =	0	x $ 18.00	$0.00	
Independent claims	1 - 3 =	0	x $ 80.00	$0.00	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ $ 270.00		
TOTAL OF ABOVE CALCULATIONS =				$860.00	
Reduction of 1/2 for filing by small entity, if applicable. A Verified Small Entity Statement must also be filed (Note 37 C.F.R. §§1.9, 1.27, 1.28).					
SUBTOTAL =				$860.00	
Processing fee of **$130.00** for furnishing the English translation later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 C.F.R. §1.492(f)).					
TOTAL NATIONAL FEE =				$860.00	
Fee for recording the enclosed assignment (37 C.F.R. §1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 C.F.R. §§3.28, 3.31). $40.00 per property.					
TOTAL FEES ENCLOSED =				$860.00	
				Amount to be refunded:	
				charged:	

a. ■ A check in the amount of $860.00 to cover the above fees is enclosed.

b. ☐ Please charge my Deposit Account No. 13-3402 in the amount of $ ______ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. ■ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 13-3402. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 C.F.R. §§1.494 or 1.495 has not been met, a petition to revive (37 C.F.R. §1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO. Customer Number 23,599

23599

PATENT TRADEMARK OFFICE

SIGNATURE

Anthony J. Zelano

NAME

27,969

REGISTRATION NUMBER

Filed: 15 MAY 2001

AJZ:kms

IN THE UNITED STATES DESIGNATED/ELECTED OFFICE

International Application No.	:	PCT/EP99/08780
International Filing Date	:	15 NOVEMBER 1999
Priority Date(s) Claimed	:	16 NOVEMBER 1998
Applicant(s) (DO/EO/US)	:	CHANDRA, Amita, et al.

Title: CRYSTALLINE POROUS SOLIDS, PRODUCTION AND USE THEREOF

PRELIMINARY AMENDMENT

Commissioner for Patents
Washington, D.C. 20231

SIR:

Although the claims of the above application were amended under Article 34 during the International Phase, applicants respectfully request that examination be based on the claims as originally filed and this Preliminary Amendment is based thereon.

Prior to calculating the national fee, and prior to examination in the National Phase of the above-identified International application, please amend as follows:

IN THE CLAIMS:

3. (Amended) The method as claimed in claim 1, characterized in that the fluid mixture has an essentially eutectic composition.

4. (Amended) The method as claimed in claim 1, characterized in that the second phase is removed in step (iii) by means of solvent extraction.

5. (Amended) The method as claimed in claim 1, characterized in that the second phase is a substance which is soluble in aqueous media.

6. (Amended) The method as claimed in claim 1, characterized in that the first phase is a water-insoluble salt.

7. (Amended) The method as claimed in claim 1, characterized in that the second phase is a water-soluble salt which is able to form a eutectic mixture with the first phase.

8. (Amended) The method as claimed in claim 1, characterized in that the first phase comprises AgCl and the second phase comprises an alkali metal halide.

10. (Amended) Porous ion-conducting solid obtainable via a method as claimed in claim 1.

14. (Amended) The use of a solid or an electrochemical cell as claimed in claim 11 as a sensor.

REMARKS

The purpose of this Preliminary Amendment is to eliminate multiple dependent claims in order to avoid the additional fee. Applicants reserve the right to reintroduce claims to canceled combined subject matter.

Respectfully submitted,

Anthony J. Zelano, Reg. No. 27,969
Attorney for Applicants
MILLEN, WHITE, ZELANO & BRANIGAN, P.C.
Arlington Courthouse Plaza 1
2200 Clarendon Boulevard, Suite 1400
Arlington, VA 22201
Direct Dial: 703-812-5311
Facsimile: 703-243-6410
Email: zelano@mwzb.com

AJZ:jmm

Filed: 15 MAY 2001

VERSION WITH MARKINGS TO SHOW CHANGES MADE

Claims 3-8, 10 and 14 have been amended as follows:

3. (Amended) The method as claimed in ~~any one of~~ claims 1~~ or 2~~, characterized in that the fluid mixture has an essentially eutectic composition.

4. (Amended) The method as claimed in ~~any one of~~ claims 1~~ to 3~~, characterized in that the second phase is removed in step (iii) by means of solvent extraction.

5. (Amended) The method as claimed in ~~any one of~~ claims 1~~ to 4~~, characterized in that the second phase is a substance which is soluble in aqueous media.

6. (Amended) The method as claimed in ~~any one of the preceding~~ claims 1, characterized in that the first phase is a water-insoluble salt.

7. (Amended) The method as claimed in ~~any one of the preceding~~ claims 1, characterized in that the second phase is a water-soluble salt which is able to form a eutectic mixture with the first phase.

8. (Amended) The method as claimed in ~~any one of the preceding~~ claims 1, characterized in that the first phase comprises AgCl and the second phase comprises an alkali metal halide.

10. (Amended) Porous ion-conducting solid obtainable via a method as claimed in ~~any one of~~ claims 1~~ to 9~~.

14. (Amended) The use of a solid or an electrochemical cell as claimed in ~~any one of~~ claims 11~~ to 13~~ as a sensor.

2/PRTS

09/831825
JC18 Rec'd PCT/PTO 15 MAY 2001

WO 00/29354 PCT/EP99/08780

CRYSTALLINE POROUS SOLIDS, PRODUCTION AND USE THEREOF

Description

The invention relates to crystalline porous solids, to a method of producing them and to the use of them.

Porous solids have attracted increasing interest in years past. They combine the characteristics of a massive solid, such as mechanical strength, with certain application-specific advantages such as e.g. reduced weight, high surface area, in some cases permeability to relatively small molecules or exclusion of relatively large molecules etc. Important fields of application for the porous materials are therefore found in separation technology or catalysis.

Previously known porous solids consist of ceramic materials having low conductivity and are produced via precursor methods or decomposition methods. They are therefore of no use or of only limited use as conductive materials (e.g. in sensor technology).

Such a method of producing porous solids having low conductivity is described e.g. in DE 41 02 430 A1. According to DE 41 02 430 A1, a fine-pored solid having a high pore volume is produced by a coarsely disperse sedimentable mixture of a liquid phase and solid particles being caused to sediment and the sediment being solidified in the presence of the liquid phase by a chemical reaction between the sediment particles to form a porous body.

DE 37 31 649 A1 describes a method of producing open-pored sintered bodies which comprises sintering a sinterable glass powder and an inorganic, soluble salt of defined granularity, whose melting point is above the melting temperature of the sinterable powder, and dissolving away, after cooling, the soluble salt

present in the sintered body. The glass powder used according to DE 37 31 649 A1 is distinguished, in particular, by high sinterability. This method too affords open-pored sintered bodies which consist of glass ceramics and have low conductivity.

There is therefore a need for porous materials having a conductivity which is higher than that of the known porous ceramics.

The present invention therefore relates to a method of producing a porous solid, characterized by the steps of:

(i) preparing a fluid mixture comprising a first phase which includes one or more inorganic ionic components, and at least one second phase, the first phase and the second phase being essentially immiscible in the solid state,

(ii) cooling the fluid mixture to a temperature below the solidification point in order to form a solid phase mixture comprising at least one first crystalline phase and a second phase, and

(iii) removing the second phase.

The advantages of the method described here consist in the simplicity of the production of porous conductive solids, which involves the preparation of a preferably eutectic mixture of at least two solid phases, at least one soluble and one insoluble phase. The morphology of the phases can be influenced by simple variation of the preparation conditions or of the quenching rate. Removal of the soluble phase results in the formation of an open pore network. Thus, porous conductive, especially ion-conducting, electroceramics are accessible which owing to the microstructure of the eutectic have high mechanical stability. The significance of the method inter alia resides in the fact that - as shown in the examples - the highly

porous materials obtained provide a large contact area and are therefore of importance for the use in sensorics, e.g. in gas sensorics. Equally, the solids according to the invention can also serve as containers for a liquid electrolyte. Owing to interface interactions, the solid can be filled in a simple manner with electrolyte liquid and subsequent bleeding of the latter (phase separation) can be prevented.

The porous solid resulting from the method has an essentially open-pored structure and, owing to its crystalline composition has a high conductivity, especially ionic conductivity. The mean pore size depends on the structure of the phase which has been dissolved away and can therefore vary over a wide range. Thus, the pores can e.g. have a size in the order of from about 20 mm to 5 μm in each spatial direction. Anisotropic pore structure can likewise be obtained, e.g. lamellar pore structures, which can have pore sizes of from 2 to 3.5 μm × 500 nm up to 1.5 μm × 20 nm to 200 nm. The degree of the porosity (fraction of the pore volume relative to the total volume) depends on the respective fractions of the first and second phases in the fluid mixture and can range from about 10 to 70%, preferably from 20 to 50%.

The fluid mixture prepared in step (i) of the method according to the invention includes at least two phases, which are miscible in the fluid state but not in the solid state. The term "fluid state" refers to a melt or alternatively e.g. a plasma. The first phase includes one or more inorganic ionic components, especially ionic compounds such as salts, for example. Preferred examples of such compounds are water-insoluble salts, e.g. silver halides, especially $AgCl$.

The second phase comprises a substance which in the solid state is immiscible with the first phase and preferably is essential miscible therewith in the fluid state. Preferably, the second phase used is a water-soluble salt compound which is able to form a eutectic mixture with the first phase. If the first phase is a silver halide, e.g. AgCl, the second phase used can be an alkaline earth metal halide or alkali metal halide, e.g. KCl, RbCl or/and CsCl. Particularly preferred is a eutectic mixture comprising about 70 mol% of AgCl and 30 mol% of KCl.

According to step (ii) the fluid mixture is cooled to a temperature below the solidification point. This affords a solid which contains a phase mixture comprising at least one first crystalline phase and a second, selectively removable phase. It is also possible for further phases to be present, which can be selectively removable soluble phases or/and insoluble phases which remain in the resulting porous solid.

Via the cooling rate it is possible to vary the morphology of the resulting solid. According to one embodiment, cooling takes place under non-segregating conditions (quenching), the cooling rate being sufficiently high to prevent crystal growth and consequently the formation of substantial crystals. In this case, the cooling rate is preferably in the range of from 10 to 50°C/min and above. In other cases, slower cooling is also possible, to enable crystal growth to the extent desired. Thus, slow cooling of a non-eutectic fluid mixture first results in a fluid eutectic composition containing particles of the first or second phase dispersed therein, which then solidifies as the temperature drops below the eutectic temperature. It is thus possible to produce a porous solid which includes two or more pore species which differ in terms of size or/and morphology.

Preferably, the fluid mixture has an essentially eutectic phase composition. Upon cooling of such a mixture, porous solids having a lamellar morphology can be obtained. The composition of the mixture is preferably in the range of ± 10 mol%, especially ± 2.5 mol% of a eutectic mixture.

The removal of the second phase from the solid can be effected, for example, by solvent extraction, if the first phase is insoluble in a particular solvent and the second phase is soluble therein. Preferentially, a second substance is used which is soluble in aqueous media (water, aqueous acid or bases). Where appropriate, however, organic solvents can also be used for the extraction.

As an alternative to a solvent extraction, the second phase can also be removed by other methods (chemical reactions or/and heating).

The invention further relates to a porous ion-conducting solid obtainable via the inventive method.

The porous solid can be employed directly for further use. Alternatively, however, it can also be ground down into smaller particles and be converted into a different shape, e.g. by compression molding. If the solid consists of an ion-conducting material it can be used in an electrochemical cell as an electrolyte, e.g. as a solid electrolyte or as a support for a liquid electrolyte. The electrochemical cell customarily contains at least two electrodes (e.g. measuring electrode and reference electrode) and the electrolyte disposed between the electrodes. The cell can be used as a sensor, e.g. as an amperometric or conductometric sensor for determining physical parameters, e.g. temperature, or chemical parameters, e.g. gaseous

substances such as H_2O, CO_2 and NH_3. By using the inventive porous solids as electrolytes, a considerable increase in the sensitivity of such sensors is possible. A porous AgCl solid is suitable, in particular, for the determination of NH_3.

The porous solid is also suitable for other applications (fluid supports, separation techniques, catalysis). For these purposes, the pores of the solid can, if required, be coated with further substances, e.g. metals, metal oxides or even biomolecules.

The invention is explained in more details by the following examples, in conjunction with the appended figures in which:

Figure 1a is a scanning electron micrograph of the lamellar structure which was obtained by cooling a fluid mixture of a eutectic composition of AgCl and KCl (30 mol% KCl, 70 mol% AgCl),

Figure 1b is a scanning electron micrograph of the porous AgCl solid obtained after the KCl phase had been dissolved away,

Figure 2a is a diagram which shows the reversible change in the conductivity in a porous AgCl solid upon switching from an Ar atmosphere to an NH_3 atmosphere and back, and

Figure 2b shows the change in the conductivity in a porous AgCl solid in the absence and the presence of a liquid electrolyte (0.5 M and 1 M $AgNO_3$) as a function of the temperature.

Examples

1. Production of a porous AgCl solid

AgCl (70 mol%) and KCl (30 mol%) are heated to 350°C in a preheated furnace. The homogeneous melt is cooled to room temperature by being removed from the furnace. Then the KCl is dissolved away by immersion in distilled water, and the resulting solid is dried in air for 24 h. A porous, mechanically stable solid is obtained. The porosity corresponds to the KCl content.

The structure of the solid before and after the extraction of KCl is shown in figures 1a and 1b.

2. Determination of NH_3

The porous AgCl solid according to example 1 is ground to a powder which is then compression-molded by uniaxial compression, using a pressure of about 30 kN/cm^2, to produce pellets having a diameter of 1 cm.

A pellet is arranged between two electrodes to produce an NH_3 sensor. Silver paste is used for the electrodes.

Figure 2a shows the change in conductivity of the porous AgCl specimen in the presence of NH_3 and inert gas (argon), respectively. A reproducible and reversible rapid change in conductivity proportional to the NH_3 concentration is observed.

3. Liquid-electrolyte support

A porous AgCl solid produced in accordance with example 1 is filled with liquid electrolyte ($AgNO_3$). Owing to the capillary forces, the liquid electrolyte is readily absorbed by the porous solid and retained therein. Figure 2b shows the change in the conductivity of a porous AgCl solid in the presence and absence of $AgNO_3$ (0.5 M and 1 M) as a function of the temperature. As

can be gathered from the diagram, the porous AgCl solid is eminently suitable as a support for liquid electrolytes.

Claims

1. A method of producing a porous solid, characterized by the steps of:
   (i) preparing a fluid mixture comprising a first phase which includes one or more inorganic ionic components, and at least one second phase, the first phase and the second phase being essentially immiscible in the solid state,
   (ii) cooling the fluid mixture to a temperature below the solidification point in order to form a solid phase mixture comprising at least one first crystalline phase and a second phase, and
   (iii) removing the second phase.

2. The method as claimed in claim 1, characterized in that the cooling is performed under non-segregating conditions.

3. The method as claimed in any one of claims 1 or 2, characterized in that the fluid mixture has an essentially eutectic composition.

4. The method as claimed in any one of claims 1 to 3, characterized in that the second phase is removed in step (iii) by means of solvent extraction.

5. The method as claimed in any one of claims 1 to 4, characterized in that the second phase is a substance which is soluble in aqueous media.

6. The method as claimed in any one of the preceding claims,
characterized in that
the first phase is a water-insoluble salt.

7. The method as claimed in any one of the preceding claims,
characterized in that
the second phase is a water-soluble salt which is able to form a eutectic mixture with the first phase.

8. The method as claimed in any one of the preceding claims,
characterized in that
the first phase comprises AgCl and the second phase comprises an alkali metal halide.

9. The method as claimed in claim 8,
characterized in that
the mixture is formed from 70 mol% of AgCl and 30 mol% of KCl.

10. Porous ion-conducting solid obtainable via a method as claimed in any one of claims 1 to 9.

11. An electrochemical cell which contains as the electrolytes a porous solid as claimed in claim 10.

12. The electrochemical cell as claimed in claim 11,
characterized in that
the pores of the solid are filled with a fluid.

13. The electrochemical cell as claimed in claim 12,
characterized in that
the fluid is a liquid electrolyte.

14. The use of a solid or an electrochemical cell as claimed in any one of claims 11 to 13 as a sensor.

15. The use as claimed in claim 14 for the determination of gases.

16. The use of a solid as claimed in claim 10 in separation technology or in catalysis.

Figure 1a

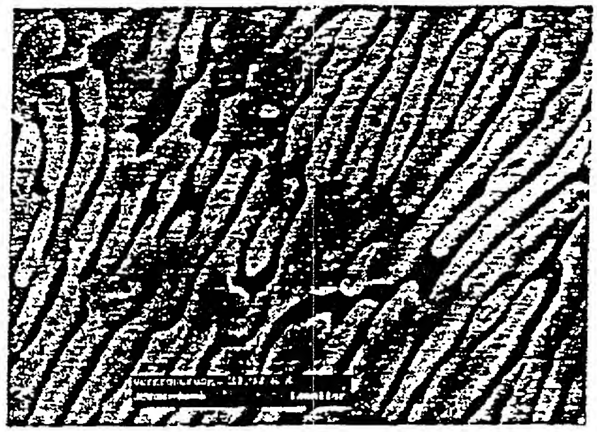


Figure 1b

Figure 2a

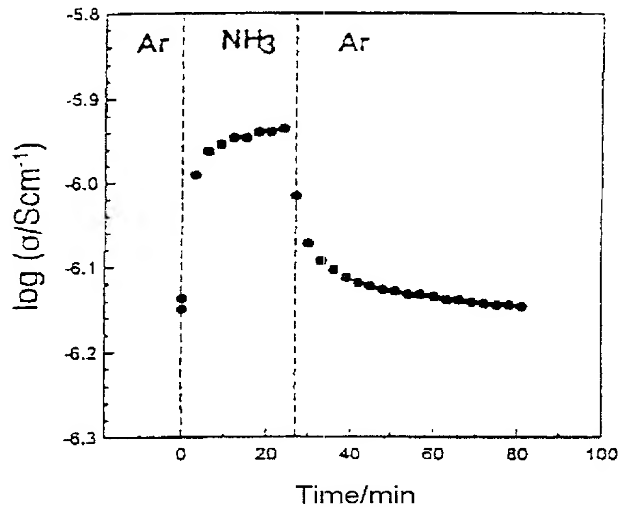
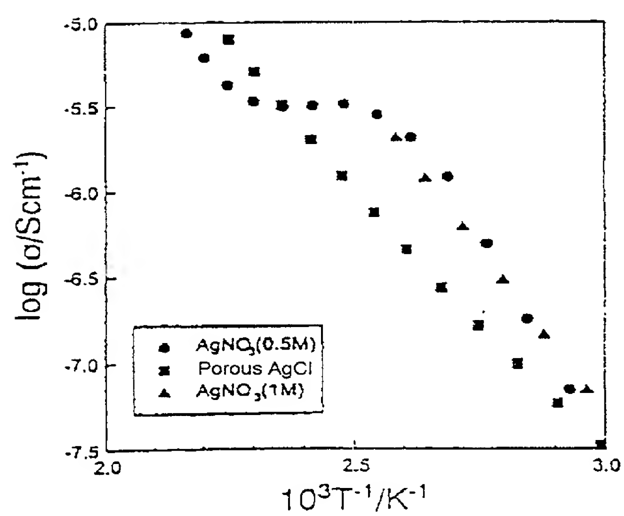


Figure 2b



Attorney Docket Number: WEICKM 10

DECLARATION FOR PATENT APPLICATION

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

CRYSTALLINE POROUS SOLIDS, PRODUCTION AND USE THEREOF

the specification of which

□ is attached hereto

■ was filed on 15 NOVEMBER 1999 as United States Application Number or PCT International Application Number PCT/EP99/08780 and (if applicable) was amended on ______

I hereby authorize our attorneys to insert the serial number assigned to this application.

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR §1.56.

I hereby claim foreign priority benefits under 35 U.S.C. §119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

PRIOR FOREIGN/PCT APPLICATION(S) AND ANY PRIORITY CLAIMS UNDER 35 USC §119			
APPLICATION NO.	COUNTRY	DAY/MONTH/YEAR FILED	PRIORITY CLAIMED
198 52 783.7	GERMANY	16 NOVEMBER 1998	YES

I hereby claim the benefit under 35 U.S.C. §119(e) of any United States provisional application(s) listed below.

PROVISIONAL APPLICATION(S) UNDER 35 U.S.C. §119(e)	
APPLICATION NUMBER	FILING DATE

I hereby claim the benefit under 35 U.S.C. §120 of any United States application, or §365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. §112.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR §1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

PRIOR U.S./PCT INTERNATIONAL APPLICATION(S) DESIGNATED FOR BENEFIT UNDER 37 U.S.C. §120		
APPLICATION NO.	FILING DATE	STATUS — PATENTED, PENDING, ABANDONED

I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected herewith: I. William Millen (19,544); John L. White (17,746); Anthony J. Zelano (27,969); Alan E.J. Branigan (20,565); John R. Moses (24,983); Harry B. Shubin (32,004); Brion P. Heaney (32,542); Richard J. Traverso (30,595); John A. Sopp (33,103); Richard M. Lebovitz (37,067); John H. Thomas (33,460); Catherine M. Joyce (40,668); Nancy J. Axelrod (44,014); James T. Moore (35,619); James E. Ruland (37,432); Jennifer J. Branigan (40,921) and Robert E. McCarthy (46,044)

Send Correspondence to Customer Number 23599

23599

PATENT TRADEMARK OFFICE

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Full Name of ~~sole or first~~ inventor (given name, family name) Amita CHANDRA	
Signature X Amita Chandra	Date X May 14, 2001
Residence Chandigarh, India INX	Citizenship India
Post Office Address Panjab University, Dept. of Physics, Chandigarh 160 014 India	
Full Name of additional joint inventor (given name, family name) Joachim MAIER	
Signature X Joachim Maier	Date X May 4 '01
Residence Wiernsheim, Germany	Citizenship Germany DEX
Post Office Address Im Kazenloch 102, D-75446 Wiernsheim, Germany	
Full Name of additional joint inventor (given name, family name) Annett SPANGENBERG	
Signature X A. Spangenberg	Date X May 09/01
Residence Stuttgart, Germany	Citizenship Germany DEX
Post Office Address Fuggerstrasse 2, D-70563 Stuttgart, Germany	
Full Name of additional joint inventor (given name, family name)	
Signature	Date
Residence	Citizenship
Post Office Address	
Full Name of additional joint inventor (given name, family name)	
Signature	Date
Residence	Citizenship
Post Office Address	

☐ Additional joint inventors are named on separately numbered sheets attached hereto.